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Multiannual Programme of the Joint Research Centre 1980-1983

1980 Annual Status Report

Plutonium fuels and actinide research

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Plutonium Fuels and Actinide Research 1980

Research staff:	117
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Projects:	1 Operation Limits of Plutonium Fuels
	1.1 Swelling of Advanced Fuels
	1.2 Oxide Fuel Transients
	1.3 Equation of State of Nuclear Materials
	2 Actinide Cycle Safety
	2.1 Formation of Actinides (FACT)
	2.2 Safe Handling of Plutonium Fuel (SHAPE)
	2.3 Aspects of the Head-End Processing of Carbide Fuel (RECARB)
	3 Actinide Research
	3.1 Crystal Chemistry
	3.2 Solid State Studies
	3.3 Applied Actinide Research

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1. Introduction

Nuclear energy is released by fission of heavy nuclei. In parallel with the fission process in a nuclear reactor, neutron capture produces elements heavier than the initial uranium. These so-called transuranium elements such as plutonium and americium have important consequences for the nuclear fuel cycle and, because of their high specific radioactivity, particularly the biologically important alpha particles which they release, must always be handled with care.

Technological aspects of the handling and behaviour of these substances during all stages of the fuel cycle, fabrication, irradiation, reprocessing and waste disposal, are thus of importance for the continued development of nuclear energy. All information should be available on the physical and chemical properties, not only of uranium compounds but also of those of plutonium and higher actinides.

Generally nuclear safety considerations focus on the potential malfunctioning of large nuclear installations. They calculate risks and establish multiple and redundant devices to minimize the probabilities of accidents. But the source of the danger is the nuclear fuel which has to be prevented from reaching uncontrolled criticality or releasing fission products and Heavy Atoms.

Thus basic research on the technology of nuclear fuels and especially those containing plutonium will always have a place in nuclear energy. Eventually the field will expand beyond plutonium because breeding and recycling will need a better knowledge also of higher elements and their compounds as to their nuclear, chemical and mechanical characteristics.

The European Institute for Transuranium Elements was established with all of the necessary facilities for this work and it serves as the focal point for a Community Research Programme,

- meeting demands not satisfied in national laboratories
- giving all Member States immediate access to this area of research
- extending research on nuclear fuels beyond the immediate needs in preparation for the future development of the cycle
- coordinating dispersed efforts in the field of basic research on actinide elements beyond the immediate interests of the nuclear industry.

By a regular exchange of ideas, experience and plans with national laboratories, the Institute has established a permanent role within the Community context, a trend which is apparent for the whole Joint Research Centre Programme of which the Plutonium and Actinide Programme is a part.

Although the general concern for reactor safety has changed the primary orientation of the programme (formerly directed towards the economy of the nuclear fuel cycle) work on the so-called "advanced" fuels continues to be a part of the Institute's activity, exercising thereby a "flywheel action" maintaining long-term research lines alive independent of the sometimes short-term fluctuations of national priorities.

2. Results

1. Operation Limits of Plutonium Fuels

Safe operation of nuclear fuel requires knowledge of the operation limits within which the specified behaviour is maintained. If these limits are for any reason exceeded it must be possible to evaluate the consequences. The swelling behaviour of advanced fuels, i. e. fuels of uranium plutonium mixed carbide or nitride (MX type) has been studied for the past 8 years and this is now nearing completion.

Oxide Fuel Transients is a new sub-project. Equation of state investigations provide the data required to evaluate the behaviour of the fuel in the hypothetical case of an uncontrolled power excursion.

1.1. Swelling of Advanced Fuels

The *objective* of this sub-project is to provide an experimentally substantiated model to describe the behaviour of fast reactor advanced fuels under the full range of normal operating conditions.

During the reporting period:

- experimental results measured in numerous irradiated fuels have been collated into a coherent picture. Two areas where information is still insufficient are a comparative study of the mechanical properties of carbide and nitride fuels based on nitride of high purity and the local fission gas analyses by transmission electron microscopy of MX-fuels in a burn-up range as large as possible in order to establish a detailed model of the fission gas kinetics, especially for temperatures below 1000 °C (Fig. 1).
- the post irradiation examination of carbide fuels from He-bonded and Na-bonded fast breeder pins of high burn-up (7 to 12 a/o) has been terminated. An empirical quantitative description of the in-pile performance of carbide fuel has been given. On this basis also carbonitride and nitride fuels may be treated for which extensive fast flux irradiation data do not exist.
- the in-pile performance of advanced fuels depends largely on their mechanical properties. Hence the fracture stress, flow stress using acoustic emission, fracture surface energy measurements and the calculation of residual stresses in pellet fragments were investigated.

1.2. Oxide Fuel Transients

Transient conditions arise from operational events such as fuel changing, load changing or maintenance requirements, all of which may be classified as normal operation. Additionally unplanned transients will inevitably occur. Examples of such transients are equipment faults or operator errors which could give rise to mismatch between coolant flow and power generation leading to excursions which are limited by the reactor safety equipment. It is necessary to evaluate the consequence of any such transient on the fuel behaviour and its subsequent life.

It is the *objective* of sub-project "Oxide Fuel Transients" to develop on a theoretical and experimental basis a physical model of the fuel response during power and temperature transients from normal operation temperature to the melting point.

Fig. 1 – Replica electron micrograph of carbide fuel section after 11 % burn-up. The larger features stem from the initial porosity, the smaller ones represent fission gas bubbles in the size range 50 to 250 nm. It is these small bubbles which cause fuel swelling.

During the reporting period,

- an irradiation programme for transient testing of fuels has been prepared.
- analysis methods have been developed to assist in interpretation of irradiation conditions.
- the post irradiation examination of transient test samples has been prepared.

Samples, which had been irradiated during 1979 in the framework of the previous sub-project "Corrosion", have been examined. Interesting results have been obtained using a micro galvanic cell to measure the oxygen potential in irradiated fuel samples. The radial gradient of oxygen potential and hence oxygen to metal ratio existing during the final days of the irradiation have been determined. This is an important result of relevance to the transient programme because physical properties of the fuel such as thermal conductivity depend on the local oxygen/metal ratio (Fig. 2).

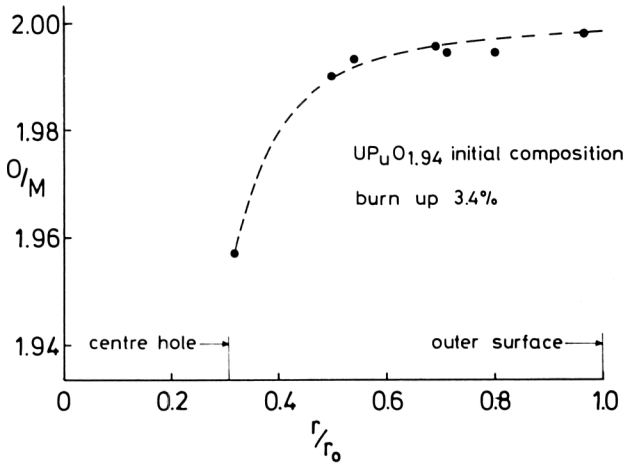

Fig. 2 - Radial variation of O/M in an irradiated mixed oxide fuel pellet.

1.3. Equation of State of Nuclear Materials

The *objective* of this work is to determine the vapour pressure generated by an irradiated nuclear fuel between 3000 K and the critical point. Initial work with UO_2 at several research laboratories has given results which vary significantly from calculations by classical thermodynamic methods (Fig. 3).

During the reporting period,

- the necessity to explain almost one order of magnitude difference in the total pressure of UO_2 at 5000 K, the experimental data being higher than the calculated pressures, involved the main effort on both the theoretical and experimental side. The theoretical activity of the project was concentrated on the study of the vaporization mechanism at extreme rates of evaporation and the gas dynamics of the Mach disk technique.
- the final objective of the studies on the mechanism of the evaporation of nuclear fuels is the determination of the ratio of the molecular species and their partial rates of evaporation under extreme conditions, which

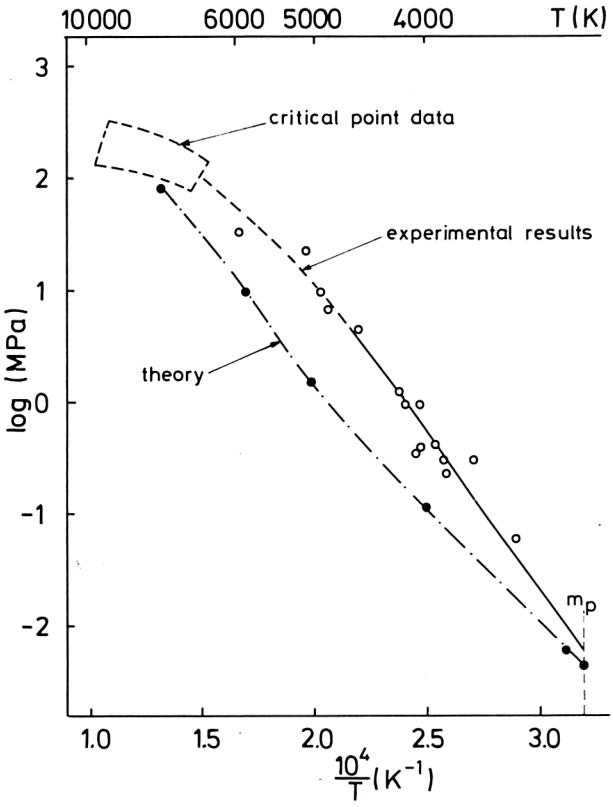

Fig. 3 - Total vapour pressure over uranium dioxide up to the critical point

leads to a departure from equilibrium rates. Above the operational limit of the Hertz-Knudsen equation, the evaporation mechanism is determined by the population density of the different energy levels. A surface model, starting from the well-defined structure of solid materials, had to be developed to determine the binding and activation energies. The suppression of the slowest rate determining reaction step in a multistep vaporization process may in principle lead to an increase in the overall evaporation rate beyond the calculated equilibrium relationship.

- mach disk measurements from 3700 K to 4200 K on UO_2, evaluated on the basis of gas dynamic relationships, confirmed the previous results from laser induced evaporation into a vacuum.

2. Actinide Cycle Safety

The safety of a nuclear fuel cycle depends among other things on the content and isotopic composition of heavy nuclides and fission products in the fuel, and on their chemical and physical form during the different stages of the cycle.

With recycling of plutonium, there is an increasing formation of actinides (FACT) heavier than plutonium. The handling risk of fuel, especially that containing recycled Pu, is enhanced (SHAPE). Fabrication of advanced fast breeder fuel poses additional problems including disposal or re-use of fabrication scrap and processing of irradiated carbide or nitride fuel (RECARB).

2.1. Formation of Actinides (FACT)

The *objectives* of this activity are

- to determine experimentally the nuclear properties of actinides irradiated in thermal and fast reactors in order to check the reliability of reactor physics calculations,
- to assess the irradiation behaviour of significant quantities of mixed uranium - minor actinide oxides considering the build-up of transplutonides by Pu recycling (Fig. 4).

During the reporting period,

- the isotope correlation technique was applied to calculate the formation of actinides,
- the preparation of irradiation experiments with americium containing fuel was started,
- mixed uranium-americium oxide fuel was prepared and characterized,
- fission yield systematics were investigated.

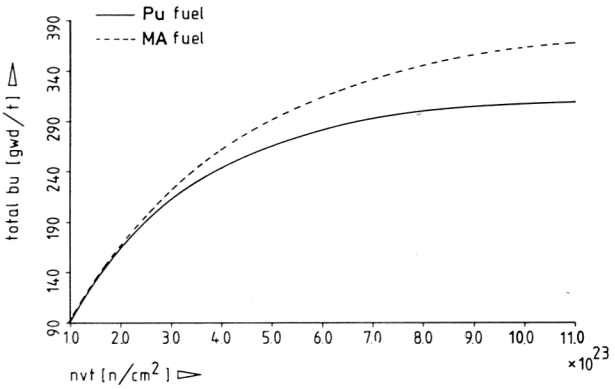

Fig. 4 - Accumulated energy (GWd/T) obtained from recycling Pu or Pu together with self-generated Minor Actinides MA (= Pu + Np + Am + Cm) in a fast reactor. (10^{23} n/cm² correspond to one fuel cycle. Out of pile time 3 years.)

2.2. Safe Handling of Plutonium Fuel (SHAPE)

As Pu will be used as ceramic (oxide, carbide, nitride ...) fuel, its handling problems are increased by the formation of aerosols. The acceptability of Pu fuels in nuclear energy depends, therefore, also on the knowledge of aerosol formation, stability and distribution as a function of fuel form, and on the availability of fabrication procedures yielding reproducible and uniform products which meet safety standards. The *objective* of this activity is to study these aspects in realistic laboratory conditions.

During the reporting period,

- an aerosol laboratory was installed,
- methods of characterization of nuclear aerosols were developed (Fig. 5),
- characterization of aerosols formed during fuel fabrication was started.

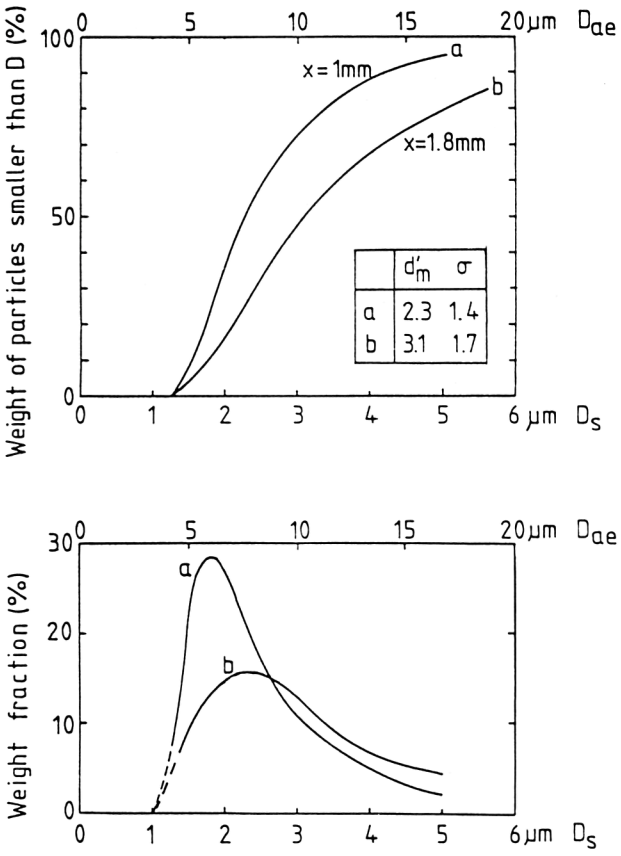

Fig. 5 - Size distribution of aerosol particles produced by vibrating $(UP_u)O_2$ pellets together at 50 cycles per second and two different amplitudes, X. As would be expected, the larger amplitude of vibration produces a slightly greater mean particle size.

D_{ae}: Aerodynamic diameter
D_S: Stokes diameter

2.3. Aspects of the Head-End Processing of Carbide Fuel (RECARB)

Future reactor fuel, irrespective of the reactor type (LWR, HTGR, FBR) or the fuel type (oxide, carbide) is expected to be reprocessed by TBP extraction after preparation of a nitric acid feed solution in a corresponding head-end process.

The *objectives* of this activity are the planned head-end process studies for advanced fuel comprising:

- basic aspects of carbide fuel (unirradiated and irradiated) oxidation and dissolution (Fig. 6),
- a comparison of different head-end reprocessing methods for advanced fuels,
- a test of a selected head-end process with an irradiated fuel pin.

During the reporting period,

- the direct dissolution of (unirradiated) MX fuel with nitric acid was studied with special emphasis on the rate of dissolution, the composition of the off-gas and of the dissolver solution,
- the main organic compounds formed during the direct dissolution of (unirradiated) carbide fuel in nitric acid were identified and their elimination by oxidation was studied.

Fig. 6 - Oxidation in CO_2 of irradiated (UPu) C for 3 hours at 700 °C. Oxide growth occurs in pores, cracks and along specific crystallographic planes within the fuel.

3. Actinide Research

The project emphasises the study of actinide metals, of intermetallics and of simple binary compounds (oxides, chalcogenides, pnictides, hydrides). The aim is to obtain information on the influence of 5f electrons and other outer electrons on different types of bonds (ionic, covalent, metallic) and to permit understanding and/or prediction of bonding-related properties. This information is obtained by well selected solid state investigations which require well characterized samples in a suitable form (e. g. single crystals, films etc.). Interpretation of the results depends on the development of bonding models on the basis of thermodynamics and solid state quantum physics in close cooperation between experimentalists and theoreticians.

The project is structured in the following activities.

3.1. Crystal Chemistry

This activity involves the preparation and characterisation of metals or compounds of known purity and structure, in a form suitable for the investigation of solid state properties. It is performed almost exclusively at the Karlsruhe Establishment.

During the reporting period,

- Pa metal was prepared for the determination of thermodynamic properties (Fig. 7),
- methods for the preparation and characterisation of (single) crystals of actinide compounds were improved,
- high pressure and low temperature X-ray diffraction techniques were applied to actinide solids.

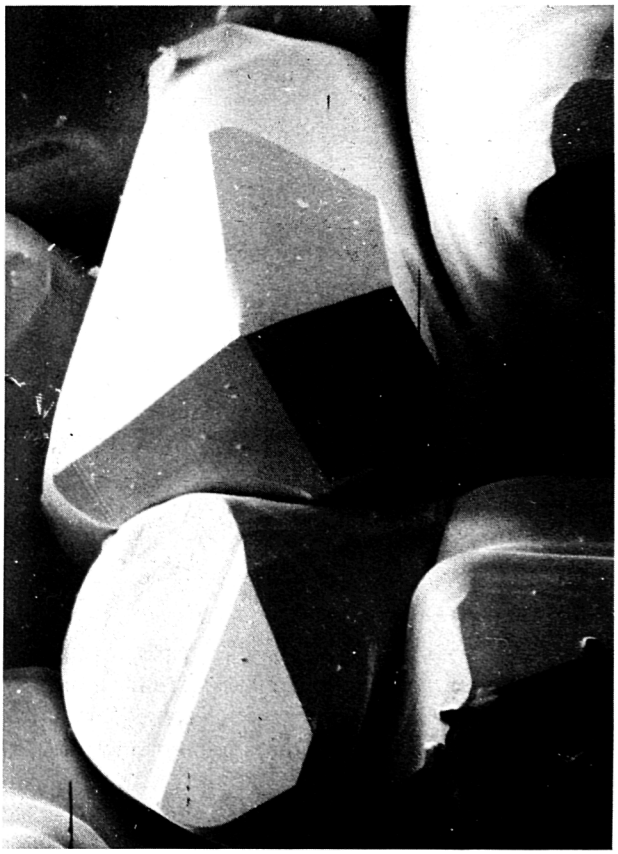

Fig. 7 - Crystals of pure Protactinium metal

3.2. Solid State Studies

Solid state properties are studied partially in collaboration with specialised laboratories within the Community.

During the reporting period, at the Karlsruhe Establishment,

- the vapour pressure of Pa metal was determined (Fig. 8),
- a thermodynamic theory developed to describe defect equilibria and transitions in oxides ("spacing statistics") was applied to specific mixed actinide oxides.
- photoelectron spectra of uranium intermetallics and uranium hydride were determined,
- band calculations using the self-consistent "linear muffin tin orbitals" (LMTO) method were compared with experimental results.

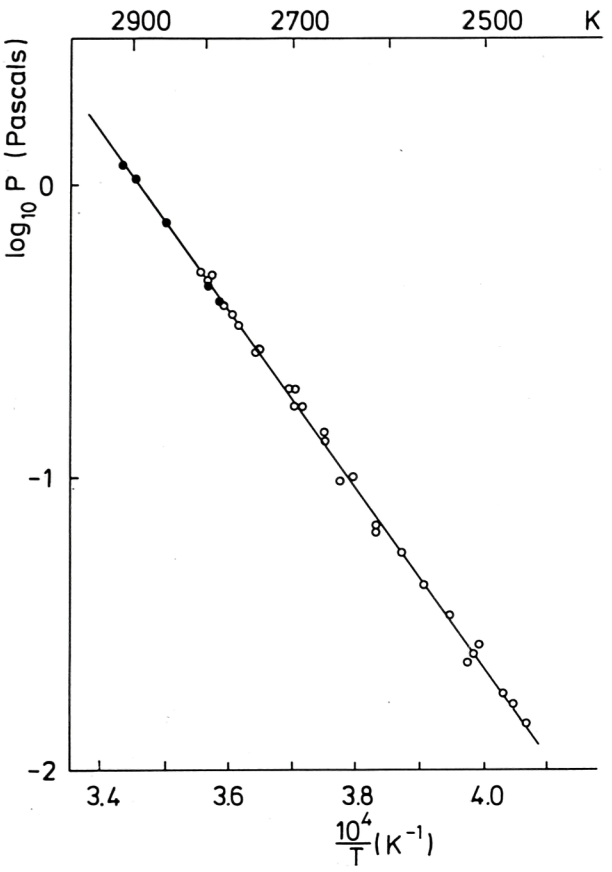

Fig. 8 – Vapour pressure of protactinium metal measured by mass effusion and mass spectrometry

3.3. Applied Actinide Research

This activity offers support to other parts of the programme, when special developments – scientific or technological – in actinide-related research are requested on a short-term-basis. It includes preparation of encapsulated actinide samples for scientific research or as radiation sources, and problems of actinide recovery and waste fixation.

Surface and catalytic properties of actinide (intermetallic) compounds will also be studied.

During the reporting period,

- the preparation of mixed actinide oxides by gel supported precipitation was attempted,
- the properties of glass and ceramics as possible matrix for solidified actinide waste were investigated.

4. Conclusions

The programme maintains, albeit at a somewhat reduced level, its involvement with the problems of advanced fast breeder fuel materials. It is a matter for speculation when these results will be needed, there is agreement that they will one day be required. At the present time problems of light water reactor safety dominate the scene, we may foresee a time when even the public media will be convinced of the safety of fast reactors and then questions of energy conservation will loom large again. The full utilisation of all fuels will be demanded and the advanced fast reactor fuels will be the subject of further development.

We are convinced that our present research is thus well justified. It has all the aspects of a Community programme, the aims are of considerable value and there is no competition with industrial interests, the joint programme has a unifying function and collaborates well with national laboratories.

Our project on safety of the nuclear fuel cycle is still only beginning. Accumulated experience in the handling of radioactive materials is being put to work and new equipment is being built up for the investigation of aerosol production and behaviour. Significant results are expected in the next few years.

Studies of the highly radioactive components of the nuclear fuel cycle have turned attention to americium and its isotopes 241 and 243. They gain increasing importance as actinide energy sources and a significant demonstration of an actinide fuel cycle seems to be possible on the basis of work done during 1980.

A closed fuel cycle involves problems of reprocessing and refabrication operations in the field of chemical engineering and ceramic process technology, which are far from being optimised. Considerable work was done in the early years of nuclear development but much less attention has been paid to them in the last decade. Our research into head end studies on carbide reprocessing technology and refabrication aspects of the higher actinides have formed a basis for further development of value to the national laboratories.

In the field of actinide research two significant advances have been made. The first is in the production of very pure materials, essentially single crystals of high quality. This is a science in itself and an absolute prerequisite of any solid state research in the field.

The second is the advanced spectroscopy in which secondary electrons are used to investigate the surface of these very pure actinide materials and, if the surface can be kept clean, the bonding properties of the bulk materials. The year 1980 saw a breakthrough in the experimental technique and we expect a number of significant results in the near future.

Research activities in the Transuranium Institute are based on excellent and in some cases unique equipment. This fact does not discharge us from delivering equivalent intellectual activity. This is also a matter of research organisation and programme management has never neglected the question of "optimum cooperation" within and between teams. An overall assessment of the research during 1980 shows an increase in productivity per person and a higher total output in spite of the ageing and unfortunate reduction of the research staff.

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